Ultra-distal fine ash occurrences of the Icelandic Askja-S Plinian eruption deposits in Southern Carpathian lakes: new age constraints on a continental scale tephrostratigraphic marker.

Kearney, R.[a,*], Albert, P.G.[a], Staff, R.A.[b], Pál, I.[c], Veres, D.[e], Magyari, E.[d], Bronk Ramsey, C.[a]

[a] Research Laboratory for Archaeology and the History of Art, School of Archaeology, University of Oxford, 1 South Parks Road, Oxford, OX1 3TG, UK.

[b] Scottish Universities Environmental Research Centre, University of Glasgow, Rankine Avenue, East Kilbride, South Lanarkshire, G75 0QF, UK.

[c] Isotope Climatology and Environmental research Center (ICER), Institute for Nuclear Research, Hungarian Academy of Sciences, H-4026 Debrecen, Bem tér 18/c, Hungary.

[d] Eötvös Lóránd University, MTA-MTM-ELTE Research Group for Paleontology, Department of Environmental and Landscape Geography, Pázmány P. stny. 1/C, H-1117 Budapest, Hungary.

[e] Romanian Academy, Institute of Speleology, Clinicilor 5, 400006 Cluj-Napoca, Romania.

*rebecca.kearney@st-annes.ox.ac.uk

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Highlights

- First cryptotephra investigation from the Southern Carpathian Mountains
- Lakes Brazi and Lia provide the most south-eastern occurrence of the Askja-S tephra
- Furthest south-eastern occurrence of an Icelandic-sourced tephra
- Improved age estimate for the Askja-S eruption of 10,824 ± 97 cal years BP
- Highlights potential of additional tephra layers to be found in this region

Abstract

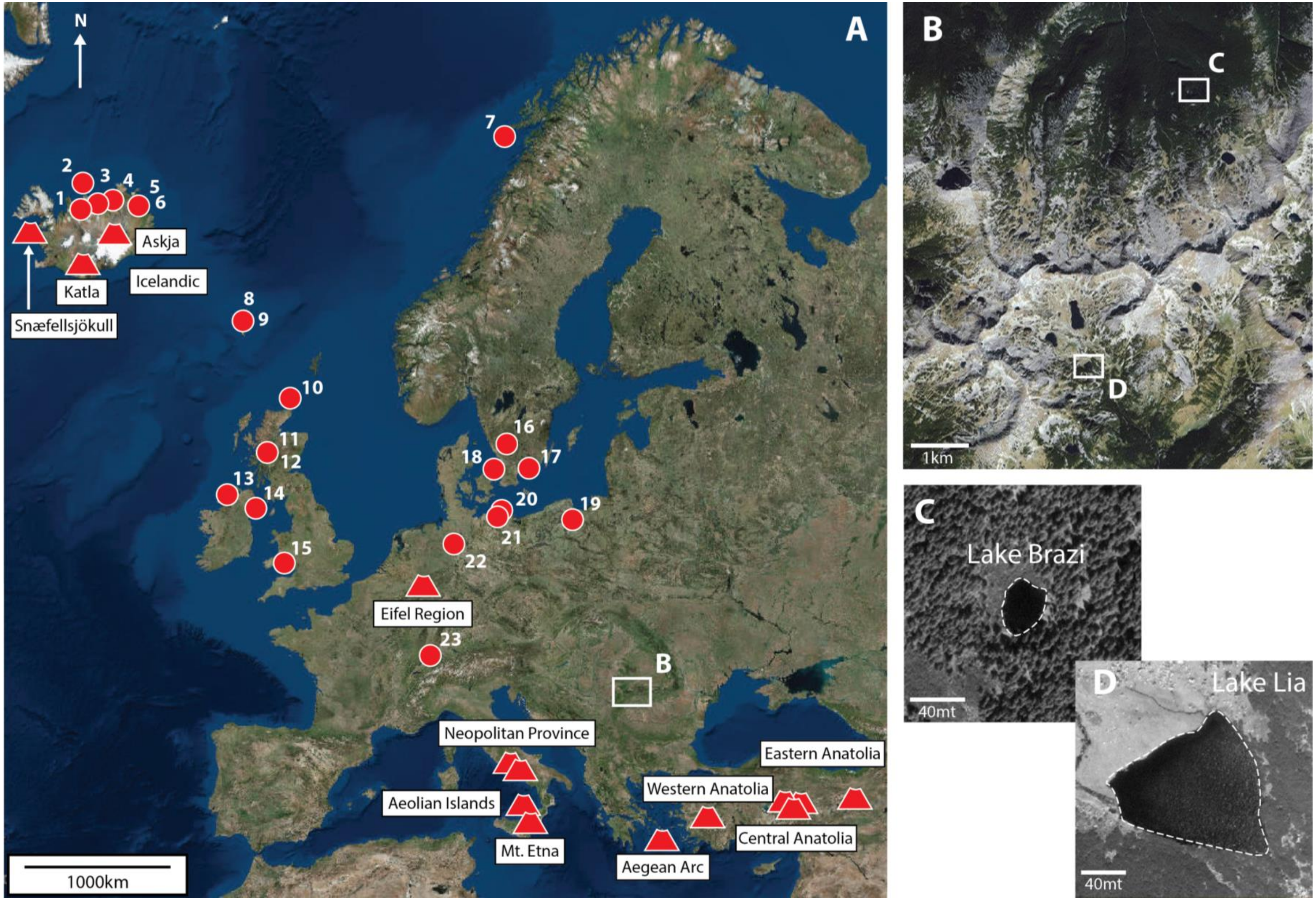
Here we present the results of the first cryptotephra investigation of two Late glacial-Holocene lake records from the Southern Carpathian Mountains in Romania, Lake Brazi and Lake Lia. The discovery of an important Icelandic tephrostratigraphic marker, the Askja-S, in the sedimentary records of both sites significantly extends the known ash dispersal from this Plinian eruption. Bayesian age-depth modelling of available radiocarbon (^{14}C) data from both sedimentary records allows us to further refine the depositional age of this ultra-distal tephra. In combination with age constraints on the tephra from other well-dated European sites, we produce an updated age for this key tephrostratigraphic marker of 10,824 ± 97 cal yrs BP (95.4% range). The Askja-S tephra is stratigraphically positioned after the palaeoenvironmental proxy response to the Preboreal Oscillation at both sites. The widespread distribution of this tephra across Europe offers the potential to assess spatio-temporal variability of this climatic signal. The discovery of the Askja-S in lake records from the Southern Carpathians highlights the likelihood of finding other ultra-distal (Icelandic) cryptotephra marker layers within the region. Additionally, given the location of the Carpathian region, it offers the opportunity to further enhance and integrate tephrostratigraphic frameworks of north-western Europe with those of the Mediterranean and Anatolia regions, which will enable a more precise comparison of palaeoenvironmental archives across Europe.

1. Introduction

The Late Glacial to Early Holocene transition (~16-8 kya) represents a period of abrupt climatic changes recorded in numerous multi-proxy palaeoenvironmental archives globally (Blockley et al., 2014; Moreno et al., 2014). These climatic changes occurred on centennial and even decadal timescales (Alley et al., 2003; Steffensen et al., 2008). Understanding the underlying driving and associated propagation mechanisms remains challenging, due to the large chronological uncertainties associated with dating methods, preventing robust inter-site comparisons of abrupt climatic transitions (Brauer et al., 2014; Bronk Ramsey et al., 2014).

The identification of widely dispersed volcanic ash (tephra) horizons can help to address these limitations (Lowe, 2011). During explosive volcanic eruptions, the fine ash component of tephra can be widely dispersed and preserved, providing time synchronous marker layers in a range of palaeoclimate records. These tephrostratigraphic markers facilitate the precise temporal comparison of disparate palaeoclimate archives (e.g. Lane et al., 2011a; Blockley et al., 2012). The identification of cryptotephra (non-visible) horizons beyond the extent of visible tephra fall layers, has enabled the application of tephrochronology over much greater areas (e.g. Jensen et al., 2014; Albert et al., 2015; van der Bilt et al., 2017). Because of this, cryptotephra horizons are becoming a powerful tool for integrating geographically disparate records, facilitating better understanding of the dynamics of abrupt climate change (e.g. Lane et al., 2013). Over the last decade, numerous European records have been subjected to detailed cryptotephra investigations, providing insights into past regional abrupt climate change, especially in northern and western Europe (e.g. Macleod et al., 2014; Lowe et al., 2015). Yet regions such as Central Eastern Europe have been largely overlooked for cryptotephra application despite the demonstrated importance of visible distal and local tephras in providing marker horizons (Veres et al., 2013; Karátson et al., 2016). However, it is expected that the region holds significant potential for successful cryptotephra investigations due to its proximity to multiple volcanic fields, which have been active during the Late Quaternary, as well as its location at the convergence of several major air circulation patterns (Fig.1A; Haliuc et al., 2017; Longman et al., 2017a, b; Obreht et al., 2017).

Figure 1. *(**A**) Landsat image (EU-DEM, 2016) of Europe showing the proximity of known active volcanic centres during the Late Glacial period and sites where the Askja-S tephra has been reported: 1) Skagafjördur (Sigvaldason, 2002); 2)* MD99-2271 (Gudmundsdóttir et al., 2011); *3) Laufas (Sigvaldason, 2002); 4) Gjastykki (Sæmundsson, 1991); 5)Bakkaflói (Norddahl and Hjort 1993); 6) Vopnafjördur (*Sigvaldson, 2002); *7) Borge Bog (Pilcher et al., 2005); 8) Høvdarhagi Bog (Lind and Wastegård., 2011); 9) Havnardalmyren (Kylander et al., 2012); 10) Quoyloo Meadow (Timms et al., 2016); 11) Inverlair (Kelly et al., 2016); 12) Turret Bank (Lowe et al., 2017); 13) Lough Nadourcan (Turney et al., 2006); 14) Long Lough (Turney et al., 2006); 15) Pant-y-Llyn (Jones et al., 2017a); 16) Mulakullegöl (Lilija et al., 2013); 17) Hässedala port (Davies et al., 2003, Wohlfarth et al., 2006); 18) Tøvelde (Larsen, 2013); 19) Lake Czechowskie (Wulf et al., 2016b); 20) Endinger Bruch (Lane et al., 2012a); 21) Lake Tiefer See (Wulf et al., 2016b); 22) Lake Hämelsee (Jones et al., 2017b); 23) Soppensee (Lane et al., 2011b). (**B**) Landsat image of the Retezat Mountains showing Lake Brazi (**C**) and Lake Lia (**D**).*



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Here, we present the important discovery of an Icelandic-derived cryptotephra, the Askja-S, within two lacustrine palaeoenvironmental records from the Southern Carpathian Mountains. This provides, for the first time, a direct tephra linkage between multi-proxy palaeoclimatic archives in south-eastern Europe with those in north-western Europe.

2. Background

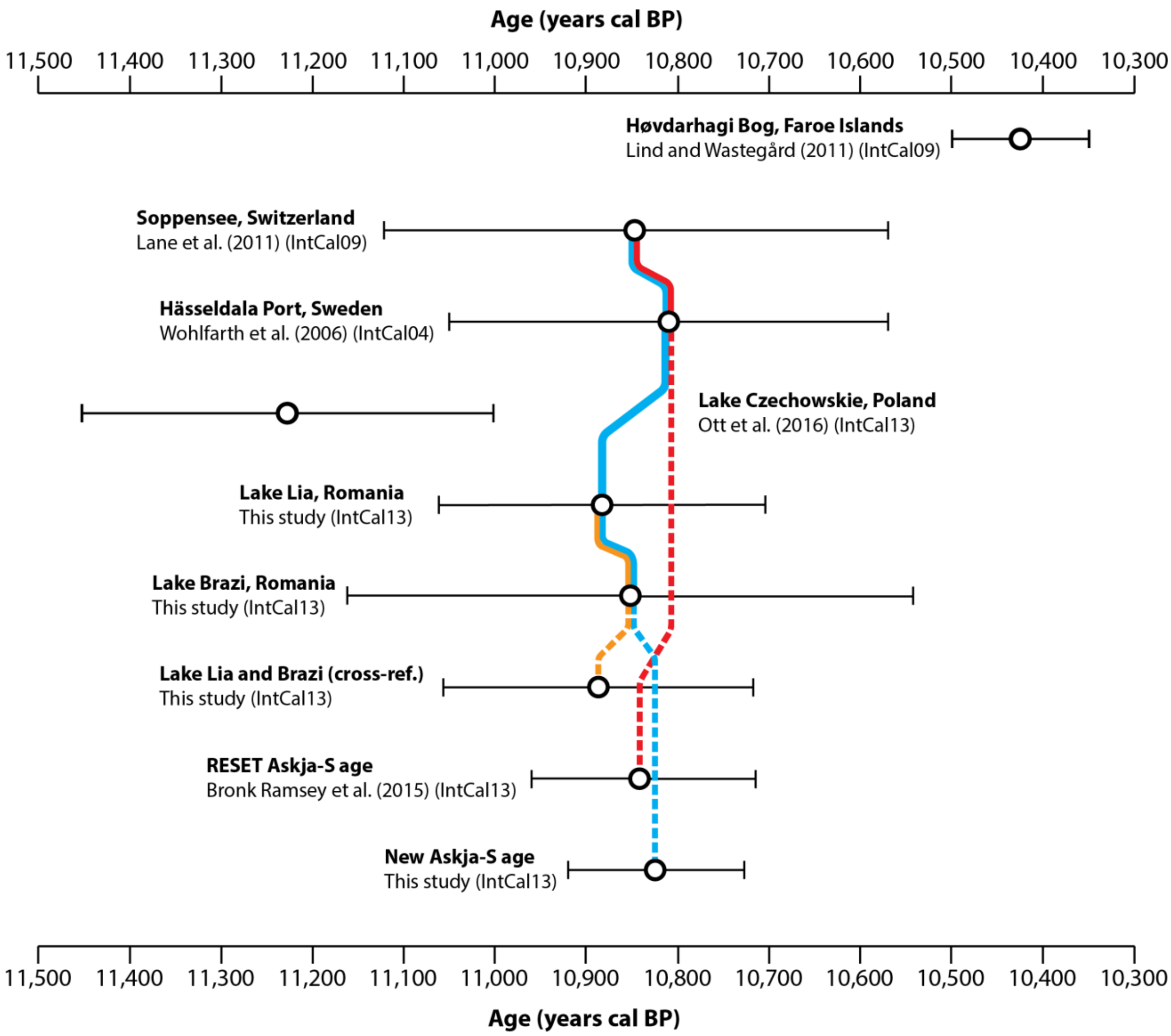
2.1. Askja-S tephra

The Askja-S tephra is the product of a Plinian eruption that formed the Askja caldera at the Dyngjuföll volcanic centre, Iceland (Sigvaldson, 2002). The estimated volume of this rhyolitic eruption is 1.5 ± 0.5 km^3 dense-rock equivalent (DRE), relative to a Volcanic Explosivity Index (VEI) of 5 (Newhall and Self, 1982; Sigvaldson, 2002). The eruption coincided with glacial retreat in Iceland after Greenland Stadial (GS)-1. Proximal Askja-S Plinian fall deposits are distributed along the north-eastern coast of Iceland (Fig.1A, Sæmundson, 1991; Norddahl and Hjort, 1994; Sigvaldson, 2002; Gudmundsdóttir et al., 2011), confirming a dominant NNE plume dispersal off Iceland.

The Askja-S was first reported distally by Davies et al. (2003) as a cryptotephra horizon in a sediment core from Hässeldala Port, Sweden. Askja-S cryptotephra has since been observed as far north as Borge, Norway (Pilcher et al., 2005), as far east as Lake Czechowskie, Poland (Wulf et al., 2016b) and as far south as Soppensee, Switzerland (Lane et al., 2011b), limiting the Askja-S dispersal to mainly north-western European sites (Fig. 1A). Most recently, the tephra has been discovered in Pant-y-Llyn, South Wales (Jones et al., 2017a).

Several ages have been suggested for the eruption of the Askja-S (Fig. 2). At Hässeldala Port, an age of 10,810 ± 240 cal yrs BP was reported by Wohlfarth et al. (2006) which has since been supported by an age of 10,846 ± 276 cal yrs BP at Soppensee (Lane et al., 2011c, Fig.2; all dates reported in this paper are at 95.4% probability). A younger age of 10,425 ± 75 cal yrs BP has been suggested by Lind and Wastegård, (2011) from Høvdarhagi Bog, Faroe Islands, whilst a significantly older age of 11,228 ± 226 cal yrs BP has been provided by Ott et al. (2016) based on a floating varve chronology from Lake Czechowskie. Arguably, the most robust age estimate for the Askja-S tephra has been produced by Bronk Ramsey et al. (2015), through integrating radiocarbon age models from multiple palaeoenvironmental records from across Europe using the RESET (RESponse of humans to abrupt Environmental Transitions) tephra lattice, producing a refined modelled age of 10,841 ± 115 cal yrs BP.

Figure 2. *Schematic representation of age estimates for the Askja-S tephra from published records, and newly remodelled data from this study. The red line indicates the sites cross-referenced for the Bayesian chronological modelling of Bronk Ramsey et al. (2015) to produce a refined the Askja-S age estimate as part of the RESET tephra lattice; the orange line shows the age estimate from modelling performed in the present study, cross-referencing of Lakes Brazi and Lia only; and the blue line indicates the records that were cross-referenced to produce our preferred the new Askja-S age building upon the RESET model by Bronk Ramsey et al. (2015) with the incorporation of the data from Lakes Brazi and Lia (this study)*



2.2. Study Sites

The Retezat Mountains form the western massif of the Southern Carpathian Mountains. This area was previously glaciated during the Last Glacial Maximum, with around 58 permanent glacial lakes still preserved (Magyari et al., 2009). Of these, two lakes, Brazi and Lia (Fig. 1B) were selected for cryptotephra investigation due to the extensive palaeoenvironmental investigations previously undertaken at both sites for the Late Glacial and Early Holocene periods (e.g. Magyari et al., 2012; Tóth et al., 2015; Buczkó et al., 2013;

Orbán et al., 2017; Vincze et al., 2017). The chronologies of both sites are based upon radiocarbon (^{14}C) dating (Magyari et al., 2009, 2012; Hubay et al., 2016; Pál et al., 2016).

Lake Brazi (45°23’47” N, 22°54’06” E, 1740m a.s.l., Fig. 1C) is located on the northern slope of the Retezat Mountains in a former kettle-hole setting (Magyari et al., 2009). Lake Lia (45°21’7.3” N, 22°52’27.0” E, 1910m a.s.l, Fig. 1D) is a small, shallow lake located 5km from Lake Brazi on the south facing slopes of the Retezat Mountains (Orbán et al., 2017; Vincze et al., 2017).

3. Methods

3.1. Tephra sample preparation

Contiguous 5cm resolution samples were taken across the sedimentologically identified Late Glacial to Early Holocene sequence of both Lake Brazi (TDB) and Lia (LIA) sediment cores (Magyari et al., 2009; Vincze et al., 2017). The glass shards were extracted following the density separation procedures described by Turney (1998) and Blockley et al. (2005) with a single modification where samples were sieved at 80µm and 15µm, rather than 80µm and 25µm. The tephra extraction residue (2.55g cm^{-3}) of each sample was mounted on to microscope slides in Canada Balsam. Volcanic glass shards were identified and counted under a high-powdered, polarizing optical microscope. Where peaks in glass shard concentration were identified at 5cm resolution, these depths were re-sampled at a higher resolution (1cm) and prepared as above.

Individual glass shards from identified peaks were concentrated and mounted on to epoxy resin stubs with the use of a micromanipulator (see Lane et al., 2014). Finally, the shards were sealed in resin, sectioned and polished for geochemical analyses.

3.2. Geochemical analysis

Major and minor element compositions of individual glass shards were measured using a JEOL-8600 wavelength-dispersive electron microprobe (WDS-EPMA) at the Research Laboratory for Archaeology and History of Art, University of Oxford. All analyses used an accelerating voltage of 15 kV, beam current of 6 nA and 10 µm-diameter beam. Peak counting times were 30 seconds for all elements apart from Na (12s), Mn (40s), Cl (50s) and P (60s). The electron microprobe was calibrated using a suite of mineral standards, and the PAP absorption correction method was used for quantification. The accuracy and precision of our analyses were assessed using analyses of the MPI-DING reference glasses (Jochum et al., 2006), which were run alongside the unknown tephra samples. Data were filtered to remove accidental analyses of minerals and biogenic silica. Volcanic glass with analytical totals <95% were also removed.

3.3. Age-depth modelling

The original radiocarbon dates from both Lake Brazi and Lake Lia (Magyari et al., 2009, 2012; Hubay et al., 2016; Pál et al., 2016) have been re-modelled using the Bayesian statistical program OxCal (version 4.3, Bronk Ramsey, 2017) applying the IntCal13 calibration curve (Reimer et al., 2013). A ‘P_Sequence’ deposition model (Bronk Ramey, 2009a) was applied for both sites utilising a variable *k* parameter to allow OxCal to independently determine the optimal variability in sedimentation rate (Bronk Ramsey and Lee, 2013). A 'general' *Outlier_Model* was applied with a 5% prior probability of any individual date being a statistical outlier (Bronk Ramsey, 2009b).

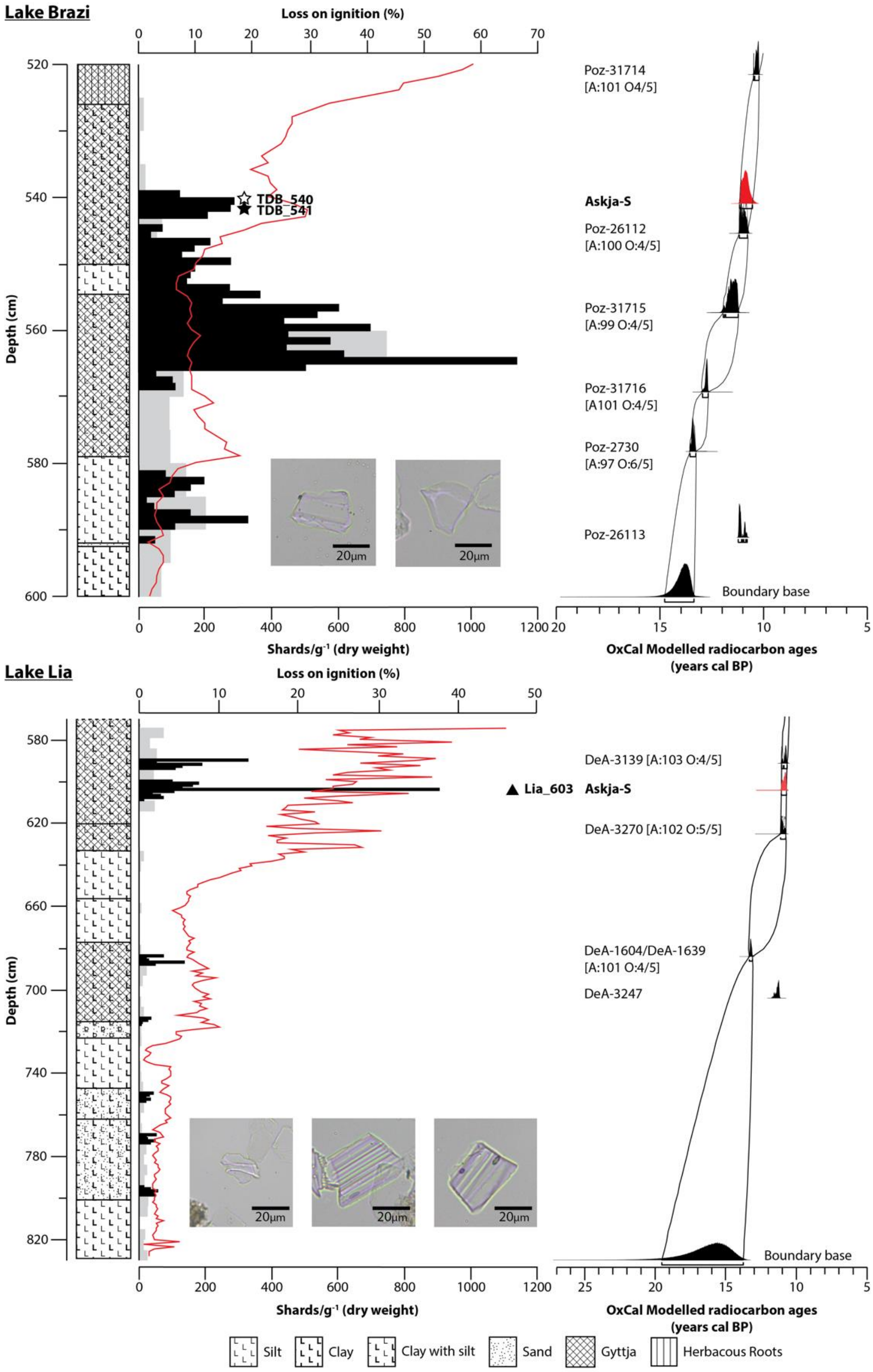
4. Results and discussion

4.1. Position and identification of cryptotephra layers

Distinct peaks in tephra shard concentration were identified in both the Lake Brazi and Lia sediment cores (Fig. 3). Robust geochemical analysis identified separate eruptions by minor and major element comparisons. For the purposes of this contribution we only present grain-specific glass geochemistry from selected peaks in glass shard concentrations in the Late glacial – early Holocene sequences of the two lakes, at 540/541 cm from the Lake Brazi sedimentary sequence (TDB_540, 288 shards g^{-1}; and TDB_541, 275 shards g^{-1}) and 603 cm in the Lake Lia record (LIA_603, 908 shards g^{-1}) (Fig. 3). Due to the nearly identical shard concentrations across TDB_540 and TDB_541, the positioning of the peak or isochron is taken as the mid-point of these depths (540.5cm). Other peaks in tephra shard concentrations, with different geochemical characteristics from the distinctive Askja-S, will be discussed in future publications.

Figure 3. *Lithostratigraphy, loss-on-ignition, total shard concentration (from low-resolution, 5cm contiguous scans, light grey; and from higher, 1cm resolution scans, black) and re-modelled age-depth models for both Lake Brazi and Lake Lia focusing on the Late Glacial and*

Early Holocene core sections. The peaks containing the Askja-S tephra that are only discussed in this paper are highlighted. Other tephra concentration peaks will be the subject of future publications. Images of individual tephra shards from these Askja-S peaks are shown.



TDB_540/541 and LIA_603 are positioned within the locally described pollen zones associated with the transition from the Younger Dryas to Early Holocene (Magyari et al., 2009, 2012; Vincze et al., 2017). The modelled age of Askja-S from Lake Brazi is 10,852 ± 309 cal yrs BP and Lake Lia 10,885 ± 183 cal yrs BP (Fig.3), within good statistical agreement of each other.

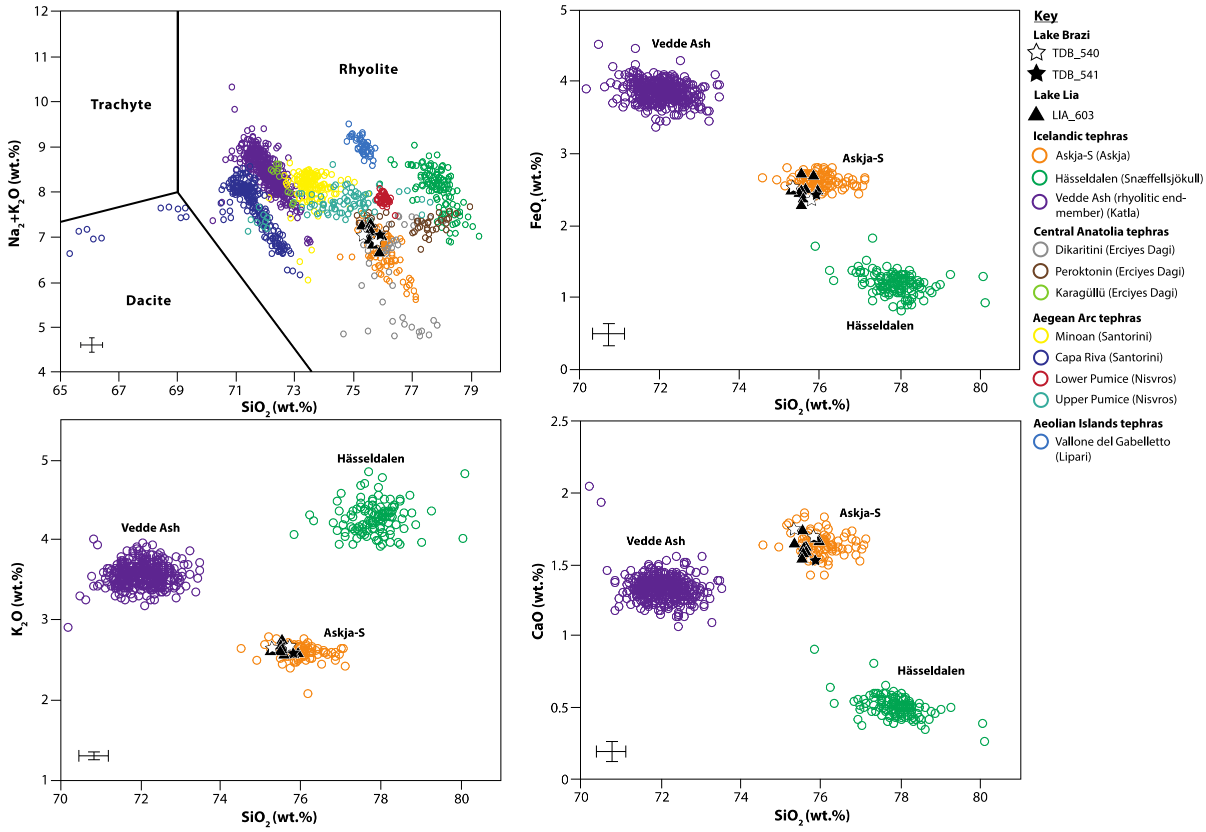
The shard morphologies of both layers exhibit a similar fluted appearance, the shards are colourless, with a maximum shard size of 20-25µm (Fig.3). The individual geochemical analyses of the glass shards in TDB_540/541 and LIA_603 are presented in Table 1, with selected major and minor element bi-plots shown in Figure 4. TDB_540/541 and LIA_603 demonstrate a homogenous, rhyolitic glass composition characterised by low K_2O (TDB_540/541: 2.60 ± 0.05 wt. %, LIA_603: 2.60 ± 0.06 wt. %; 1 s.d.) and high CaO (TDB_540/541: 1.65 ± 0.11 wt. %; LIA_603: 1.61 ± 0.06 wt %; 1 s.d.). Due to this distinctive glass chemistry and the chronostratigraphic position of the TDB_540/541 and LIA_603 samples, we can confidently correlate them to the Askja-S erupted from the Dyngjuföll volcanic centre, Iceland (Sigvaldson, 2002) as well as to identified distal occurrences of ash from this eruption at multiple sites across north–western Europe (Davies et al., 2003; Pilcher et al., 2005; Lane et al., 2011b; Wulf et al., 2016b; Jones et al., 2017b) (Fig 1A, Fig 4). The ultra-distal discovery of the Askja-S tephra, over 3000 km from volcanic source in the Southern Carpathian Mountains dramatically extends the known dispersal of fine ash (typically < 25 µm glass shards) from this Plinian eruption and, furthermore, represents the first identification of an Icelandic tephra in south-eastern Europe.

Table 1. *Major element glass compositions (normalised to 100%), along with raw (non-normalised) analytical totals of individual tephra shards from Lake Brazi (TDB_540, TDB_541) and Lake Lia (LIA_603). Unnormalised data can be seen in Supplementary Material A.*

Sample	SiO_2	TiO_2	Al_2O_3	FeO_t	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	Cl	Analytical Total
TDB_540	75.35	0.34	12.37	2.63	0.16	0.26	1.73	4.42	2.61	0.05	0.08	95.89
	75.82	0.3	12.54	2.47	0.06	0.24	1.68	4.17	2.64	0.03	0.05	98.08
TDB_541	75.93	0.34	12.23	2.52	0.12	0.23	1.53	4.48	2.54	0.01	0.07	97.04
Mean	*75.7*	*0.33*	*12.38*	*2.54*	*0.11*	*0.24*	*1.65*	*4.36*	*2.6*	*0.03*	*0.07*	*97*
SD (1σ)	*0.31*	*0.02*	*0.15*	*0.08*	*0.05*	*0.02*	*0.11*	*0.16*	*0.05*	*0.02*	*0.01*	*1.09*
LIA_603	75.56	0.34	12.49	2.6	0.16	0.25	1.52	4.26	2.63	0.07	0.11	98.58
	75.32	0.29	12.42	2.61	0.14	0.27	1.63	4.63	2.59	0.04	0.08	98.48
	75.66	0.28	12.56	2.62	0.09	0.25	1.61	4.26	2.54	0.05	0.08	97.27
	75.53	0.27	12.41	2.58	0.05	0.29	1.72	4.4	2.59	0.07	0.09	99.3
	75.59	0.26	12.48	2.41	0.1	0.26	1.57	4.5	2.74	0.02	0.06	98.3
	75.9	0.29	12.28	2.79	0.13	0.24	1.66	4.09	2.53	0.01	0.08	99.94
	75.6	0.3	12.21	2.82	0.06	0.21	1.6	4.49	2.62	0.02	0.08	98.73
	75.63	0.33	12.36	2.49	0.16	0.24	1.58	4.47	2.61	0.06	0.07	99.87
	75.98	0.27	12.01	2.62	0.07	0.27	1.65	4.41	2.55	0.08	0.1	98.74
Mean	*75.64*	*0.29*	*12.36*	*2.62*	*0.11*	*0.25*	*1.61*	*4.39*	*2.6*	*0.05*	*0.08*	*98.8*
SD (1σ)	*0.2*	*0.03*	*0.17*	*0.13*	*0.04*	*0.02*	*0.06*	*0.16*	*0.06*	*0.03*	*0.02*	*0.82*

Figure 4. *Geochemical bi-plots of the major elemental data from Lake Brazi and Lake Lia with known rhyolitic eruptions during the Late Glacial to Early Holocene from volcanic sources close to the sites. Reference data for tephras are from Central Anatolia (Hamann et al., 2010, Cullen et al., 2014, Tomlinson et al., 2015), Aegean Arc (Eastwood et al., 1999, Kwiecien et al., 2008, Tomlinson et al., 2012, 2015, Cullen et al., 2014), Aeolian Islands (Albert et al., 2017) and Icelandic tephra (Vedde Ash: Birks et al., 1996, Wastegård et al., 1998, 2000, Turney et al., 1997, 2006, Raner et al., 2005, Pilcher et al., 2005, Davies et al., 2005, Blockley et al., 2007, Lane et al., 2011b, c, d, 2012a, b; Hässeldalen: Lane et al., 2012b, Housley et al., 2013, Davies et al., 2003, Lind and Wastegård, 2011, Liljn et al., 2013, Larsen and Noe-Nygaard., 2014, Wulf et al., 2016b; and Askja-S: Sigvaldason 2002, Davies et al., 2003, Turney et al., 2006, Lane et al., 2011b, Lind and Wastegard, 2011, Wulf et al.,*

2016, Kelly et al., 2016, Jones et al., 2017b, Lowe et al., 2017). Error bars presented are 2 standard deviations of repeat analysis of the StHs6/80-G MPI-DING standard glass.



4.2. Chronological modelling

Owing to the occurrence of the Askja-S in both lake records, its eruption age can be subsequently refined using cross-referencing in OxCal (Bronk Ramsey, 2009a), producing an updated age of 10,889 ± 174 cal yrs BP for the deposition of this tephra within both lake records. This is within statistical agreement with the Askja-S age proposed by Bronk Ramsey et al. (2015), but contradicts the older and younger ages reported by Ott et al. (2016) and Lind and Wastegård (2011), respectively. Whilst other, younger tephras from Askja volcano have been reported (e.g. the Askja-L and Askja-H; Jóhannsdóttir, 2007, Striberger et al., 2012, Gudmundsdóttir et al., 2016), there is no evidence to date of these tephras having been identified beyond Iceland. Therefore, we presume that the divergent ages presented by Ott et al. (2016) and Lind and Wastegård (2011) are the result of problematic site chronologies, rather than the misattribution of these tephra layers.

Combining the chronological modelling of the Askja-S tephra in Lakes Brazi and Lia with the data from the other two sites, Soppensee and Hässeldala Port whose ages for the tephra are within statistical agreement with our own, improves upon the age estimate of the RESET tephra lattice by Bronk Ramsey et al. (2015, Fig.2). The new results from the model provide a refined age of 10,824 ± 97 cal yrs BP (Fig.2).

5. Future research

4.3. Palaeoenvironments

The discovery of an Icelandic tephra layer in two palaeoenvironmental records from the Southern Carpathians provides a key tephrostratigraphic linkage between western and northern Europe with south-eastern Europe (Fig. 1). Here it is particularly important as the Askja-S tephra is chrono-stratigraphically positioned in the Early Holocene close to where several abrupt climatic transitions have been reported, for instance the Preboreal Oscillation (PBO, 11.5-11.4 b2k,) and the '10.7 kya event' which are identified in numerous European palaeoclimate records (Björck et al., 1997; Wohlfarth et al., 2006; Lowe et al., 2015; Ott et al., 2016). However, these events remain somewhat poorly understood in terms of their environmental impacts and geographical expression (e.g. Björck et al., 1996, 1997; Wohlfath et al., 2006). Both lakes, have undergone high-resolution, multi-proxy palaeoenvironmental investigations across this time period (e.g. Magyari et al., 2012; Buczkó et al., 2013; Tóth et al., 2015, in press; Pál et al., 2016, in press, Finsinger et al., 2014, in press; Vincze et al., 2017). The additional identification of the Askja-S within both records, provides an ideal opportunity to investigate comparative environmental dynamics and duration of the PBO through improving chronological modelling from north-western Europe to the Southern Carpathians.

4.4. Expansion of tephrostratigraphic frameworks

This finding demonstrates that the Askja-S tephra provides a powerful tephrostratigraphic marker extending from north-west Europe to south-eastern Europe. The identification of this cryptotephra horizon is not uniform across north-west European sites that have undergone detailed cryptotephra investigations, revealing a patchy distribution of this ash fall event (Jones et al., 2017a).

A number of factors could be responsible for the distal occurrences of this widespread ash dispersal from Iceland. Following an initial NNE plume dispersal off Iceland, evident in proximal deposits (Sæmundsson, 1991; Norddahl and Hjort 1993; Sigvaldason, 2002), the resulting complex distribution of ash fall evidenced across Europe may have been governed by the variable/evolving concomitant weather patterns, or different atmospheric circulation at changing heights in the plume. Modern Icelandic eruptions have exhibited complicated dispersals following their initial dispersals away from Iceland, seemingly affected by these factors (e.g. Davies et al., 2010; Gudmundsson et al., 2012).

The 15 µm sieve mesh size used here was crucial to the retention and identification of the finest grain ash particles produced during this eruption (<25 µm). Typically in previous studies a 25 µm mesh was employed and may be responsible for the Askja-S fine-grain material not being recovered by the cryptotephra procedures employed. Therefore, our findings provide encouragement for re-investigations of the temporally relevant sediments at these sites. Speculatively, the concentration of Askja-S tephra in the Lake Brazi and Lia records could have been influenced by orographic precipitation induced by the Carpathian mountain range. Additional factors such as regional bias of cryptotephra investigations (Watson et al., 2017), site-specific sedimentary taphonomic processes (Pyne-O'Donnell, 2011) and lack of continuous high-resolution cryptotephra searches (Timms et al., 2016) may all have acted to limit the currently known spatial distribution of this tephrostratigraphic marker in Europe.

Crucially, the identification of Icelandic tephra in Lake Brazi and Lake Lia, which are geographically closer to other productive volcanic regions (e.g. Italian, Aegean and Anatolian volcanoes), highlights the enormous potential of these sites for the future integration of regional tephrostratigraphic frameworks from multiple volcanic sources.

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Supplementary material A: Raw geochemical data

Supplementary material B: OxCal coding

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